Ultrafast LIPSS: from coherence to dynamic evolution

J.-P. Colombier[1], H. Zhang[1,2], E. Bévillon[1], C. Li[1,3], X. Sedao[1], C. Maurice[4], S. Reynaud[1], N. Faure[1], G. Cheng[3], F. Garrelie[1], F. Pigeon[1] , R. Stoian[1]

1- Université de Lyon, F-42023, France, CNRS, UMR5516, Laboratoire Hubert Curien, Université Jean Monnet, 42000 St-Etienne, France
2- Laser Micro/Nano Fabrication Laboratory, School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China
3- State Key Laboratory of Transient Optics and Photonics, Xian Institute of Optics and Precision Mechanics, CAS, 710119 Xian, Shaanxi, China
4- Ecole Nationale Supérieure des Mines de Saint-Etienne, Laboratoire Georges Friedel, CNRS, UMR5307, 42023 St-Etienne, France

E-mail: jean.philippe.colombier@univ-st-etienne.fr

Ultrafast light coupling with solid surfaces shows strong potential for nanostructuring applications, relying on the capacity to localize energy on a wide range of materials and to design periodic patterns involving transient matter transformations. Laser-Induced Surface Structures (LIPSS) consisting of regular periodic self-organized surface nanopatterns are often supposed to result from surface plasmon excitation, while the onset of periodic self-arrangement of matter is also observed in non-plasmonic materials. To shed a new light on this unsettling discrepancy, we have conducted a comprehension study devoted to define the excitation mechanisms from two perspectives, one related to the electromagnetic origin of spatial optical enhancement and one to the transient optical and thermo-mechanical response of the irradiated material.

To identify the electromagnetic origin of localized photoexcitation arrangements, 3-dimensional Finite-Difference Time-Domain calculations have been carried out on semi-conductors and metals [1]. An initially random distributed rough surface reveals that LIPSS can be initiated by the coherent superposition of far-field and near-field scattered waves and the refracted waves, potentially involving collective excitation on plasmonic materials. This indicates that a complex electromagnetic origin, mainly based on the coherence of the laser field, predicts a spatially-ordered energy deposition. The electromagnetic structured field highlights the role of the evolving surface topology on the pulse-to-pulse development of the pattern [2]. The transient optical response is then a second crucial question to identify the mechanisms responsible for spatial optical resonances. Under conditions of electron-phonon nonequilibrium [3], we have investigated by both *ab initio* calculations and pump-probe experiments the optical behavior of tungsten material, which is a typically non-plasmonic metal in infrared regime, potentially switching to a surface plasmonic state during ultrafast irradiation. Subtle effects on the electronic structure suggest that transient variation of optical properties can be large, impacting the surface response and fostering the collective electron charge excitation.

Finally, the localized sub-surface region experiencing high photoexcitation has shown to undergo phase transformation and thermo-mechanical change at the nanoscale [4]. Dedicated electron backscatter diffraction experiments and transmission electron microscopy reveal that lattice defects are distributed inhomogeneously, depending on the surface crystal orientation. These are a signature of the resulting transient thermodynamic states following ultrafast laser energy absorption, with practical relevance on the feedback effects dependent on the different relaxation ways, *i.e.* defects formation or surface growing.

[1] H. Zhang, J. P. Colombier, C. Li, N. Faure, G. Cheng, and R. Stoian, Coherence in ultrafast laser-induced periodic surface structures, Phys. Rev. B, vol. 92, 174109 (2015).
[2] C. Li, G. Cheng, J. P. Colombier, N. Faure, S. Reynaud, H. Zhang, D. Jamon, and R. Stoian, Impact of evolving surface nanoscale topologies in femtosecond laser structuring of Ni-based superalloy CMSX-4, J. Opt., vol. 18 (1), 015402 (2016).
[3] E. Bévillon, J.P. Colombier, V. Recoules, and R. Stoian, Free-electron properties of metals under ultrafast laser-induced electron-phonon nonequilibrium: A first-principles study, Phys. Rev. B, vol. 89, 115117 (2014).
[4] X. Sedao, C. Maurice, F. Garrelie, J.P. Colombier, S. Reynaud, R. Quey, F. Pigeon, Influence of crystal orientation on the formation of femtosecond laser-induced periodic surface structures and lattice defects accumulation, Appl. Phys. Lett., vol. 104 (17), 171605 (2014).